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Variation of both tritium (^3H) and beryllium (^7Be) concentrations in air, rain and humidity samples collected at Ioannina, North-western Greece

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Abstract Studies concerning hydrological cycle, the movement of water through the atmosphere, surface and underground but also the ocean circulation, often use radioactive isotopes with short or long half lives to evaluate the time scales of these processes. The study of the temporal and spatial distribution of such radioactive isotopes help to understand the water movement. Usually these isotopes are produced in the upper atmosphere from the interaction of cosmic rays with nuclei of atmospheric gases. Among these isotopes, tritium (^3H) and beryllium (^7Be) are very important due to their comparatively short half lives, 12.33 y for ^3H and 53.12 d for ^7Be . Observed annual fluctuations of the production and consequently the transportation and precipitation of the isotopes on the earth surface, can be attributed to the climate conditions of the troposphere and stratosphere and to the seasonal changes of the boundaries between them.

In the present work results of a study of the variations of ^3H and ^7Be concentrations in air, liquefied air humidity and precipitation are presented. Tritium in precipitation and humidity were measured using liquid scintillation counting, while ^7Be in air and rain samples with gamma spectroscopy. An overall trend of decreasing concentrations of ^7Be during the winter months is observed showing a reversed behavior during the first spring months. Concentrations of ^7Be in rain samples were for the majority of the cases, below the limit of detection. Tritium concentrations in rain similarly to those measured in humidity, tend to increase during the winter months and decrease during spring.

Keywords Tritium, ^7Be , LSC, gamma spectroscopy

INTRODUCTION

Tritium (^3H) and beryllium (^7Be) are radioactive isotopes produced in the upper atmosphere from the interaction of cosmic rays with nuclei of atmospheric gases. Due to their comparatively short half lives, 12.33 y for ^3H and 53.12 d for ^7Be , they are used as tracers of the water cycling in the atmosphere, surface and underground waters and also atmospheric and ocean circulation. An annual fluctuation of the production and consequently the transfer and precipitation of the isotopes on the earth surface is observed which can be attributed to the climate conditions of the upper and lower parts of the atmosphere and also to the special seasonal changes of the boundaries between these two parts of the atmosphere, i.e. troposphere and stratosphere. Natural levels of ^3H concentrations in the atmospheric

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precipitation vary from 0.5 to 2 BqL⁻¹. The Tritium Laboratory of the Archaeometry Center of the University of Ioannina is among the few laboratories in Greece, which can perform tritium measurements in water samples [1,2,3,4]. A series of tritium concentration measurements in precipitation samples from the area of Ioannina, North-western Greece, are presented in Fig. 1. Beryllium-7 is measured in filters where an amount of air (usually 4-6·10³ m³) passes through and the air particles, where ⁷Be atoms are attached, are collected. Filters are placed into petri dishes and are measured by means of γ ray spectroscopy. In the present work the preliminary results of an ongoing study of the variation of both ³H and ⁷Be concentrations in air, rain and humidity samples collected at Ioannina, North-western Greece are presented.

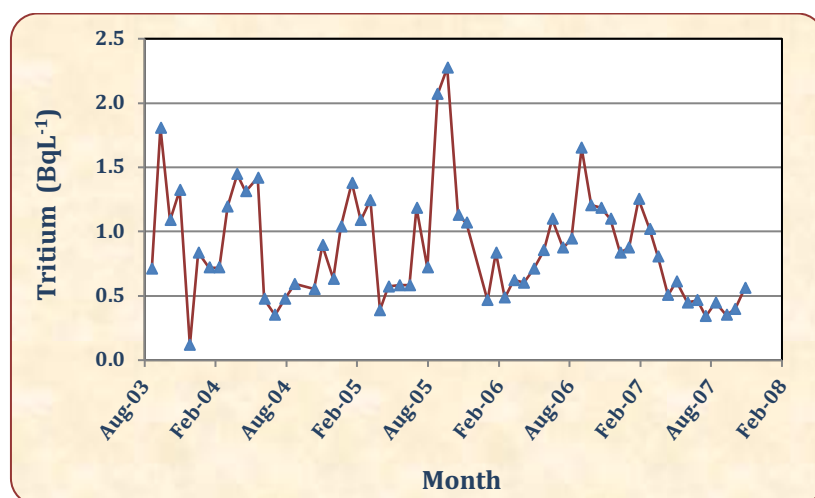


Figure 1. Tritium concentration in precipitation during 2003- 2008, at Ioannina.

MATERIALS AND METHODS

During a seven months period from (October–2017 to April-2018), 21 air samples were collected on glass-fiber filters (20x25 cm²), for 3-4 days sampling time and with air flow of 1 m³min⁻¹. Thirty six (36) rain samples were collected during storm events at the same period and were measured for tritium concentration as well for ⁷Be. Finally 19 condensed humidity samples were collected during the same storm events to measure both ³H and ⁷Be concentrations. Tritium in rain and humidity samples was determined using liquid scintillation counting, while for ⁷Be in air samples collected in filters and rain samples γ spectroscopy was utilized. All samples were measured for ³H and ⁷Be at the Archaeometry Center of the University of Ioannina. The Center is equipped with a super low-level background, liquid scintillation analyzer Tri-Carb 3170TR/SL (PerkinElmer, Inc), which is capable to measure very low concentrations of ³H for applications in radiation protection and underground water measurements. Without any further processing, 8 mL of each sample were added in a low potassium glass vial of 20 mL capacity and mixed with 12 mL of Ultima Gold LLT scintillation cocktail, suitable for low level ³H measurements. Each sample was measured for 1400 min. Rain and humidity samples were placed in plastic containers of 275 ml volume and were measured for ⁷Be content with a HPGe broad energy detector with 1.9

keV FWHM at 1.33 ^{60}Co photo-peak and 26% relative efficiency. Air filters were measured for ^7Be content also in the same detector.

| <i>Month</i> | Tritium | | Beryllium-7 | | |
|-----------------|--------------------------------------|--|--|--------------------------------------|----------------------------|
| | <i>Rain</i> (BqL^{-1}) | <i>Humidity</i> (BqL^{-1}) | <i>Air filters</i> (mBqm^{-3}) | <i>Rain</i> (BqL^{-1}) | <i>Rain Height</i> (mm) |
| Oct-2017 | 1.19 ± 0.04 | 1.06 ± 0.04 | 4.19 ± 1.58 | - | 20.5 |
| Nov-2017 | 1.23 ± 0.33 | 1.16 ± 0.29 | 3.58 ± 1.19 | 0.81 ± 0.9 | 73.6 |
| Dec-2017 | 1.31 ± 0.05 | 0.87 ± 0.03 | 1.7 ± 0.19 | 0.42 ± 0.38 | 104.6 |
| Jan-2017 | 1.53 ± 0.54 | 1.3 ± 0.04 | 1.67 ± 1.22 | 0.48 ± 0.6 | 18.2 |
| Feb-2017 | 1.47 ± 0.24 | 1.55 ± 0.46 | 0.6 ± 0.17 | 0.11 ± 0.28 | 66.6 |
| Mar-2017 | 1.31 ± 0.65 | 0.94 ± 0.35 | 1.32 ± 0.52 | 0.55 ± 0.65 | 61.6 |
| Apr-2017 | 2.46 ± 0.69 | 1.52 ± 0.05 | 2.95 ± 1.47 | - | 4.8 |

Table 1. Tritium and Beryllium monthly concentrations in rain and humidity during the period from October-2017 to April-2018, at Ioannina

RESULTS AND DISCUSSION

The results are presented in detail in Fig. 2 and 3 and monthly mean values with the associated uncertainty in Table 1. Beryllium concentrations in the air samples, show an overall trend of decreasing during the winter months while there is a small but obvious increase of the concentrations during the first spring months.

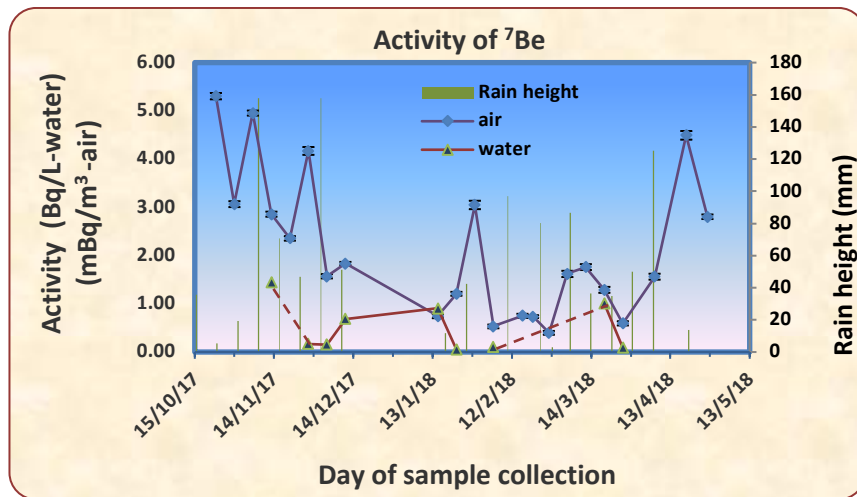


Figure 2 Beryllium-7 concentration in air and rain samples during the period from Oct-2017 to Apr-2018, at Ioannina. Weekly rain height (mm, second axis) is also reported

A similar trend was reported in another study in Thessaloniki, Northern Greece [5]. Concentrations of ^7Be in rain samples were in the majority of the cases, below the limit of detection. In some cases values were still low but measurable. Tritium concentrations in rain were similar to those measured in humidity samples and generally tend to increase during the whole period of the study. Similar results considering ^3H concentrations in rain have been

observed in previous studies [2, 3]. This variation in tritium concentrations is also observed in Fig. 1, where data from a 5-years period are presented. A weak correlation was found between the temporal evolution of ^7Be and ^3H concentrations. This may be confirmed when more results will be obtained. Tritium monthly mean concentrations in rain water varied from 1.2 - 2.5 BqL $^{-1}$ showing an average value of 1.5 ± 0.5 BqL $^{-1}$, while monthly averages of humidity samples varied from 0.9 - 1.5 BqL $^{-1}$ with an average of 1.2 ± 0.4 BqL $^{-1}$. Individual ^3H monthly concentration means with the associated uncertainties ($\pm 1\text{sd}$) are presented in Table 1. Beryllium monthly concentration means in the air were found to vary from 0.6-4.2 mBqm $^{-3}$, with an average value of 2.2 ± 1.5 mBqm $^{-3}$ while in the rain the values were very low varying from 0.1 - 0.8 BqL $^{-1}$. Also individual ^7Be monthly concentration means with the associated uncertainties ($\pm 1\text{sd}$) in air filters and rain are presented in Table 1. Meteorological data for the rain height (mm) are presented graphically in Fig. 2 and 3 (weekly values) and in Table 1 (monthly total). It is noticeable that ^7Be concentrations in the air tend to decrease significantly after a strong storm event while tend to increase during periods of drought.

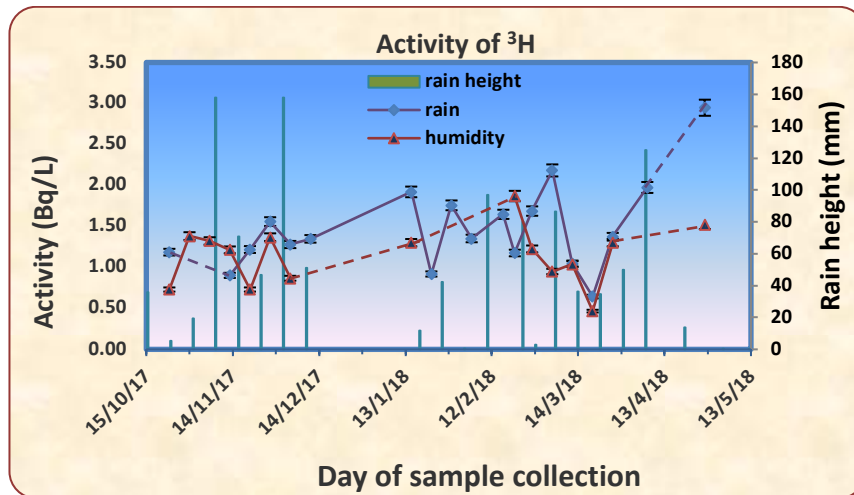


Figure 3 Tritium concentration in rain and humidity during the period from Oct-2017 to Apr-2018, at Ioannina. Weekly rain height (mm, second axis) is also reported

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